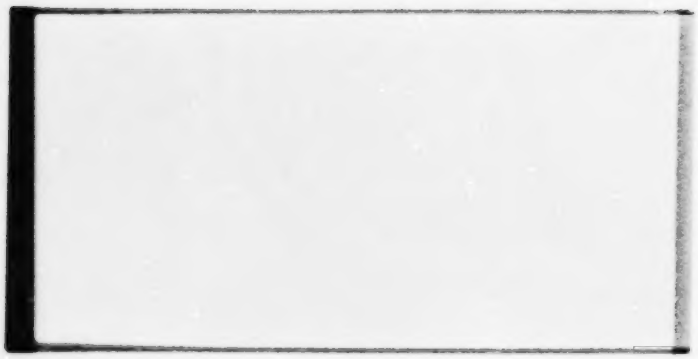
ontario hydro
research division



Named in honour of Dr. W.P. Dobson, the Research Division's director from 1914 to 1953, the building shown on the front cover was first occupied in 1961. Along with the adjacent pump testing complex and the high-voltage, high-current and radioactive materials laboratories, it accommodates much of the research, development and testing work performed in support of the diverse technical needs of Ontario Hydro.

The Research Division's staff is engaged on a wide range of problems relating to the design, construction, operation and maintenance of the power system and to the efficient, economical use of electric power.

Ontario Hydro, Research Division
800 Kipling Avenue,
Toronto, Ontario, Canada
M8Z 5S4

Telex: 06-984-525

Phone: (416)231-4111

91602
new (82-10

COMPLIMENTARY COPY
Please Credit Ontario Hydro Research if
Reproduced in Whole or in Part.

ontario hydro
research division

TRITIUM UPTAKE AND DESORPTION KINETICS IN TISSUE-FREE WATER FRACTIONS OF TOMATO PLANTS
- FINAL REPORT -

Report No 83-33-K

F.S. Spencer
Biologist
Biological Research Section
Chemical Research Department

ABSTRACT

The kinetics of tritiated water (HTO) uptake and desorption in tissue-free water fractions of the foliage and fruit of tomato (*Lycopersicon esculentem*, cv vendor) were determined in controlled environments. Most uptake data followed first-order kinetics, fitting the equation $C_t = C_\infty (1-e^{-kt})$, where C_t is the HTO concentration at time t in pCi/ml of water, C_∞ the steady-state concentration, and k the uptake rate constant in hr^{-1}. Similarly, most desorption data fitted the equation $C_t = C_i (e^{-kt})$ where C_i is the initial HTO concentration and k is the desorption rate constant. During atmospheric HTO exposure and clean water irrigation the uptake rate constants were 0.112 ± 0.045 (95% confidence interval) to 0.708 ± 0.288 hr^{-1} for foliage and 0.060 ± 0.037 hr^{-1} for green fruit. During HTO irrigation, the rate constants were smaller, 0.035 ± 0.004 hr^{-1} for foliage and 0.023 ± 0.006 hr^{-1} for green fruit. The steady-state concentration ratios during atmospheric exposure were 0.45 for foliage/atmosphere but only about one-half this ratio for green fruit/atmosphere (0.30) and ripe fruit/atmosphere (0.19). During root irrigation with HTO the foliage/soil water ratios were 0.70 to 0.83 and the green fruit/soil water ratios were 0.77 to 0.82. No significant change in HTO concentration occurred in ripe fruit following harvest. During replacement of tritiated irrigation water by clean water the desorption rate constants were 0.056 ± 0.004 hr^{-1} for foliage and 0.004 ± 0.001 hr^{-1} for green fruit. The green fruit HTO concentration decrease may have been due to dilution by clean water during growth rather than HTO desorption.

job	file	date	report no.
740634-305-490	837.88	March 11, 1983	83-33-K

TABLE OF CONTENTS

	Page
INTRODUCTION	1
METHODS	2
1) Controlled Environment Chamber Operating Conditions	2
2) Plant Stock and Growing Conditions	4
3) Monitoring and Analytic Methods	5
(a) Measurement of HTO Concentration in Chamber Atmosphere	5
(b) Chamber Monitoring of Temperature, RH, CO_2 and Light Intensity	5
(c) Sampling of Foliage and Fruit	6
(d) Extraction of Tritiated Water from Samples and Measurement of Radioactivity	6
(e) Conditions for HTO Desorption from Harvested Fruit	6
4) Data Analysis and Quality Assurance	6
RESULTS AND DISCUSSION	8
1) Controlled Environment Chamber Performance: Tritium Uptake Kinetics	8
2) HTO Uptake in Tomato Fruit and Foliage during Atmospheric Exposure to Tritiated Water	9
3) HTO Uptake in Tomato Fruit and Foliage during Irrigation with Tritiated Water	10
4) HTO Concentration Ratios of Tomato Fruit and Foliage Relative to Exposure Source	19
5) HTO Desorption from Harvested Ripe Tomato Fruit	21
6) On-the-Vine HTO Concentration Changes in Tomato Fruit and Foliage	25

TABLE OF CONTENTS (cont'd)

	Page
CONCLUSIONS	27
REFERENCES	31
APPENDICES	33
DISTRIBUTION	last page

LIST OF FIGURES

		Page
Figure 1	Controlled Environment Chamber Layout	3
Figure 2	HTO Uptake in Tomato Fruit and Foliage during Atmospheric Exposure to Tritiated Water Vapour with Roots Isolated from Atmosphere	10
Figure 3	HTO Uptake in Tomato Fruit and Foliage during Exposure to Tritiated Water Vapour with Roots Exposed to Atmosphere	12
Figure 4	Tomato Foliage HTO Uptake during Exposure to a Steady-State Concentration of Tritiated Water Vapour with Roots Exposed to Atmosphere	15
Figure 5	HTO Uptake in Tomato during Irrigation with Tritiated Water and Roots Isolated from Atmosphere	17
Figure 6	HTO Desorption in Harvested Tomato Fruit	22
Figure 7	Fresh Weight Loss after Tomato Fruit Harvest	24
Figure 8	On-the-Vine Changes in HTO Concentrations of Fruit and Foliage when HTO Irrigation was followed by Clean Water.	26

LIST OF TABLES

		Page
Table I	HTO Uptake Kinetics in Tomato Fruit and Foliage during Exposure to Tritiated Water Vapour	13
Table II	HTO Uptake Kinetics in Tomato Fruit and Foliage during Irrigation with Tritiated Water	18
Table III	HTO Concentration Ratios of Tomato Fruit and Foliage Relative to Exposure Source	20
Table IV	HTO Desorption Kinetics in Tomato Fruit and Foliage when HTO Irrigation was followed by Clean Water.	28

LIST OF APPENDICES

Appendix IA	Controlled Environment Chamber HTO Uptake Characteristics for Low-Level Exposures	33
Appendix IB	Controlled Environment Chamber HTO Uptake Characteristics for High-Level Exposures	34
Appendix II	HTO Collection Efficiency with a Two-Impinger Train	35
Appendix III	Effect of Sampling Time on Impinger Performance	36
Appendix IV	Comparison of Atmospheric Comdensate and Impinger Air Sampling for Measurement of Chamber HTO Concentration	37

ontario hydro
research division

To Mr. H.N. Isaac
Manager
Nuclear Mtls, Mgnt Dept

TRITIUM UPTAKE AND DESORPTION KINETICS IN TISSUE-FREE WATER FRACTIONS OF TOMATO PLANTS - FINAL REPORT -

INTRODUCTION

Tritium, the only radioactive isotope of hydrogen, is produced in CANDU pressurized heavy water reactors by the interaction of neutrons with deuterium in the moderator and heat transport systems. Present CANDU reactor operation results in the release of tritium to the environment primarily as tritiated water, HTO, in both the vapour and liquid states. Future tritium recovery systems installed at CANDU reactor sites will result in the release of elemental tritium, HT/T_2, as well as HTO. Although these releases are low and well below accepted health and safety limits, they, together with world-wide nuclear power operations constitute an important source of environmental tritium. Other tritium sources include that produced by nuclear weapons testing currently limited by treaty, and that formed naturally by the effects of cosmic radiation on nitrogen and oxygen in the upper atmosphere (Quisenberry 1979, Eisenbud *et al* 1979).

Research was undertaken, initially at the request of the Nuclear Materials Management Department in 1979, to develop a better understanding of tritium behaviour in vegetation surrounding CANDU nuclear facilities. Specifically, tritiated water (HTO) uptake and desorption kinetics in horticultural species was investigated because of interest in the utilization of moderator cooling water as a waste-heat application in commercial greenhouses (see interim report, Ont Hyd Res Div Report 80-52-K). This final report contains the results of further experiments dealing with the uptake and desorption of HTO into tissue-free water fractions of tomato fruit and foliage as well as the basic information presented in the interim report. Two sets of experimental conditions have been employed in these studies, one where plants were exposed to atmospheric tritiated water vapour and roots irrigated with clean water and the second where plants were irrigated with tritiated water.

The information contained in this report is directed towards the development of a predictive model to describe the environmental behaviour of tritium, in particular of pathways leading to man, and to permit more definitive radiological assessment of tritium.

METHODS

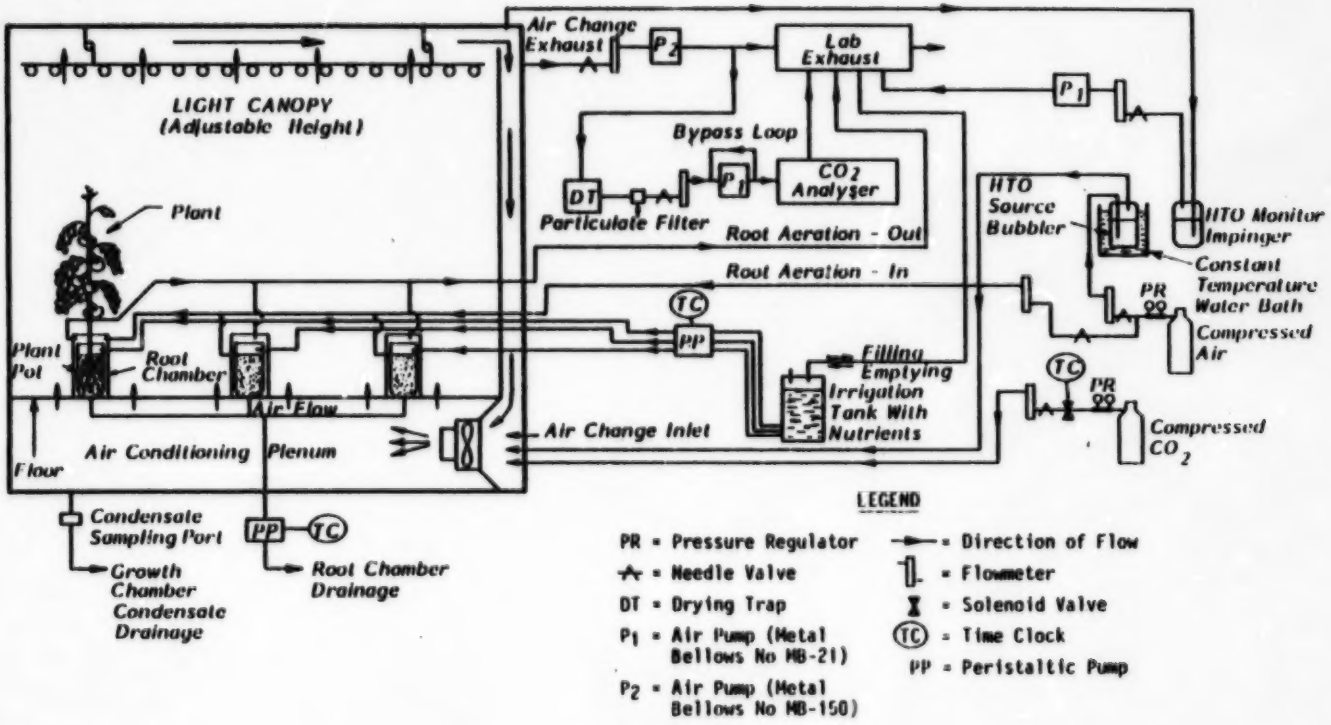
1) Controlled Environment Chamber Operating Conditions

Plants were exposed to tritiated water (HTO) in a commercially manufactured Conviron Model E 15 growth chamber. In each experiment six tomato plants were grown on a floor area of 85 cm x 190 cm; height was 180 cm. The light canopy was adjusted to keep it approximately 30 cm from the top of the plant canopy (Figure 1). Illumination was provided by 16 Sylvania VHO cool-white 183 cm fluorescent lamps and 12 Sylvania 60 W incandescent bulbs, which were spaced to provide uniform intensity in the chamber. At a distance 30 cm below the light canopy the intensity was 650 to 725 $\mu E/cm^2/sec$ and at 60 cm it was 300 to 360 $\mu E/cm^2/sec$ with plants present in the chamber.

In certain experiments with isolated root systems, tomato shoots (stem, foliage and fruit) were isolated from the roots to provide independent HTO exposure of shoots by HTO vapour in the chamber atmosphere or of roots by irrigation with tritiated water. In experiments designated 'exposed' the root systems were not contained in isolation chambers. Root isolation chambers were constructed of 3 mm thick, clear, acrylic, cylindrical side-walls and 13 mm thick end-covers, thereby enclosing each plant pot. The tomato stem protruded through a 2 cm diameter opening in the top cover which was sealed with lanolin paste.

Irrigation, root aeration, exhaust and drainage lines entered the chamber through tapped connections, and the covers sealed with self-amalgamating polyisobutylene tape, so that there was virtually no exchange of air or HTO vapour between the root and foliar environments. This was confirmed by measurement of HTO levels in the root chamber drainage during atmospheric fumigation; the values were similar to the background HTO levels, about 7.5 pCi/ml of water.

HTO was introduced into the growth chamber as tritiated water vapour for the foliar and fruit uptake experiments. In early experiments (Experiments 1 and 4) a stream of dry compressed air at a rate of 100 cc/min was passed through tritiated water of 29 to 32 µci/ml in a gas washing bottle, maintained at 30°C in a water bath. Complete saturation of this air stream was assured and thus a delivery rate of 98.5 nCi/min was introduced in the growth chamber (Figure 1). In later experiments (Experiments 2 and 3) the gas washing bottle tritium concentration was raised to 125 µCi/ml and the flow rate adjusted to 75 cc/min. The bath temperature was lowered to 15°C to prevent potential condensation in the delivery lines. For root uptake experiments, the irrigation water containing nutrients was spiked with 5 mCi/ml tritiated water to provide a 18,380 pCi/ml solution (Experiment 5) and 20,420 pCi/ml (Expt 6). In both atmospheric exposure and root irrigation experiments HTO source solutions were prepared from 5 mCi/ml HTO purchased from Amersham Corp.



187722 RD

FIGURE 1

CONTROLLED ENVIRONMENT CHAMBER LAYOUT

(Diagram shows only 3 of 6 root chambers and irrigation lines utilized)

Air flow within the chamber was provided by two sub-floor fans which provided uniform vertical velocity of about 0.5 m per second. In each experiment with atmospheric tritium exposure, air was exhausted from the chamber at 17 L/min; this provided about one air change every two hours, enough to prevent the build-up of phytotoxic gases such as ethylene but low enough to avoid unnecessary HTO release to the atmosphere. With this low air change rate, it was necessary to supply CO_2 during the day photoperiod to replenish that consumed during photosynthesis; a 35 cc/min delivery rate of 100% carbon dioxide was sufficient to maintain a concentration of 400 ± 25 mm^3/m^3, which was 25 to 50 mm^3/m^3 above the average ambient concentration in our laboratory.

In tritiated water irrigation experiments the growth chamber air change rate was about 23 changes/hr, the maximum rate. This rate was used to minimize tritium air concentrations.

2) Plant Stock and Growing Conditions

Tomato seeds, (*Lycopersicon esculentum*, cultivar vendor) were germinated in flats and seedlings transplanted in 8 L plastic pots containing Redi-Earth, a commercial mixture of equal amounts of peat and vermiculite with starter fertilizer. In Experiments 1, 4 and 5 seedlings were propagated using 20-20-20 fertilizer (Ontario Ministry of Agriculture and Food recommendations, Publication No 365) (20% ea of nitrogen, phosphorous and potassium expressed as N, P_2O_5 and K_2O) in a simulated waste-heat greenhouse at Pickering. In all other experiments (2, 3 and 6) seedlings were grown in the Research Division greenhouse using modified Hoagland's nutrient solution adjusted to pH 6.5 (Laughans, 1978). Upon transfer from the greenhouse to the growth chamber, plants were acclimated to the experimental exposure conditions (see below) for a minimum of 24 hours, except in experiment 3 where plants were introduced directly into the growth chamber containing a steady-state concentration of HTO vapour, after acclimation to laboratory conditions (23°C, ∿50% RH, 60 $\mu E/m^2/sec$ light intensity).

Irrigation was carried out in the growth chamber twice daily, on the first and eighth hour of a 16-hr day photoperiod, with half-strength Hoagland's solution. On each occasion 670 mls of nutrient were applied by peristaltic pump. Root aeration, in isolated root experiments, was accomplished by feeding 17 cc/min dry compressed air into each isolation chamber which had independent exhaust lines (Figure 1).

The daylight portion of the photoperiod was sixteen hours, during which the chamber temperature was maintained at 23 ± 1°C and relative humidity at 70 ± 2%, giving a vapour pressure deficit of 0.85 kPa across the leaf surface, (ie the vapour pressure difference between the inside and outside of the leaf, assuming 100% relative humidity inside). During the eight-hour dark period, temperature was lowered to 18 ± 1°C to simulate greenhouse conditions and reduce plant respiration, and relative humidity was reduced to 62 ± 1% to maintain the same vapour pressure deficit as during the day.

3) Monitoring and Analytic Methods

(a) Measurement of HTO Concentration in Chamber Atmosphere

The concentration of tritium in the water vapour of the growth chamber atmosphere was monitored by one of two methods in this report. In the first report atmospheric condensate, which formed at a steady rate of about 1 L/hr due to the rapid alternation of cooling/heating and dehumidification/humidification cycles, was collected for analysis by liquid scintillation counting (Ontario Hydro Research Division Report 80-52-K).

In the second method, 2,500 cc of chamber air containing tritiated water vapour were collected in a two-impinger sampling train containing liquid scintillation cocktail. Two 125 ml glass impingers, each containing 50 mls of cocktail, PCS II, were used to collect 2,500 cc of chamber air. Air was drawn through the impingers for 5 minutes by a Metal Bellows MB-21 pump at a flow rate of 500 cc/min controlled by a calibrated Matheson 602 rotameter. At room temperature ($\sim$21°C) collection efficiency of the first impinger relative to total tritium activity collected in two impingers was shown to be over 96% for sampling periods of 3 to 20 minutes (Appendix II). For a 5 minute collection period, as routinely performed in this study, the collection efficiency of the first impinger was 99.2%. The relationship of total tritium activity collected in two impingers was demonstrated to be proportional to the sampling time over the 3 to 20 minute period with slight decline at 30 minutes (Appendix III). Small quantities of particulate matter in the growth chamber atmosphere, removable by a glass wool filter, were not found to affect collection performance for periods of 3 to 20 minutes. This filter was not used subsequently in routine chamber air monitoring.

(b) Chamber Monitoring of Temperature, Relative Humidity, CO_2 and Light Intensity

Chamber temperature and relative humidity were monitored continuously with a built-in dry bulb/wet bulb recorder, and these measurements were calibrated periodically with an Ulbrecht Model 761 aspirated psychrometer. Light intensity was measured with a Lambda Instruments Model LI-185A meter using a quantum sensor to provide photosynthetically active radiation values. A Beckman Model 864 infrared analyzer and Philips 25 cm strip-chart recorder continuously analyzed carbon dioxide concentrations in the growth chamber. This analyzer was calibrated twice weekly using nitrogen zero gas and 275 and 500 mm^3/m^3 carbon dioxide span gases. All gas flow rates were measured with Matheson 600 series flow meters calibrated *in situ* using soap film manometers.

(c) Sampling of Foliage and Fruit

Foliage and fruit were sampled randomly for tritium analyses from six plants during each experiment, except for the uppermost (upper or apical quarter) young foliage which was left intact to supply photosynthate for growing fruit. Older lower foliage, if clearly senescent, was not sampled.

(d) Extraction of Tritiated Water from Samples and Measurement of Radioactivity

Tissue-free water tritium in plant samples (fruit, foliage and roots) was quantitatively extracted with an azeotropic toluene water mixture (bp = 85°C) to provide 5 to 10 ml of tissue-free water (Moghissi *et al* 1973). Approximately 30 to 50 g fresh weight of plant material were extracted with 2.5 to 3.0 volumes of liquid-scintillation grade toluene. Controls were run using 30 ml of well water containing less than 170 pCi/L of water. Tissue-free water samples were analyzed for radioactivity in the Research Division's Radioactive Materials Laboratory. Routinely, one ml aliquots of each sample were mixed with 14 ml of PCS II Amersham scintillation cocktail and counted in a Searle MkIII liquid scintillation counter.

(e) Conditions for HTO Desorption from Harvested Fruit

Desorption of harvested fruit (Figure 6) took place in an air-conditioned laboratory having about seven air changes per hour. The room temperature was 24 ± 1°C, while relative humidity varied from 36 to 56%, as monitored by a Bristol Model F4069 thermohumidigraph calibrated with an Ulbrecht Model 761 psychrometer.

4) Data Analysis and Quality Assurance

Condensate and foliage sampling were not replicated in these experiments because sample variation was typically less than 1% of mean values. Rather, emphasis was placed on more frequent sampling during uptake and desorption time courses. Subsampling of individual fruit also yielded small standard deviations, typically 1 to 5% of means, whereas variation between fruit on the same plant was 20 and 40% of means. Therefore, fruit sampling was generally triplicated. It should be noted that variation in sample counting (machine error) based on occasional triplicate subsampling was always very small, typically standard deviations were less than 1% of means, (data not shown).

The pattern of tritiated water uptake in foliage, fruit and chamber atmosphere with time was found in most experiments to follow first-order kinetics, ie uptake was proportional to concentration. The following equation was used to represent these uptake relationships:

$$C_t = C_\infty (1 - e^{-kt}) \qquad (1)$$

where

C_t = tritium concentration at time t, expressed in pCi/ml of water

C_∞ = steady-state concentration (asymptotic value) reached when $dC/dt = 0$

t = time in hours

k = initial uptake rate constant at $t \approx 0$, expressed in hr^{-1}

$t_{1/2}$ = 0.693/k, the time required to reach one-half of the steady-state concentration, expressed in hours

In each instance where the above relationship was found to be valid, the uptake versus time curves were computer fitted by a non-linear least squares method (NLIN procedure of Statistical Analyst Systems, SAS Institute, Cary, NC). The same computer program was used to estimate C_∞ and k values from which the initial uptake rate $C_\infty k$, and the time to reach 50% of the steady-state concentration ($t_{1/2}$=0.693k) were calculated. Similarly, desorption kinetics were found in certain instances (Figure 8) to follow first-order relationships, $C_t = C_i (e^{-kt})$, where C_i = initial tritium concentration expressed in pCi/ml of water and k = desorption rate constant. Desorption curves following this pattern were computer fitted as above.

On two occasions quality assurance checks were carried out between our laboratory and Safety Services, Pickering. On each occasion there was very good agreement for atmospheric condensate and fruit free-water samples, as shown below. Values shown are the means ± two standard deviations (2σ) of either two (Safety Services data) or three (our data) independent measurements of one sample.

Sample Type	Research Data (pCi/ml)	Pickering Data (pCi/ml)
1. Atmospheric condensate	3468±86	3488±10
2. Fruit free-water	355±4	350±1
3. Well water	7.6±3.7	<0.170

The detection limit in our laboratory was 7 to 8 pCi/ml compared to about 0.17 pCi/ml at Pickering, accounting for the lack of agreement in the well-water sample. This well-water, used in distillation controls, and our laboratory distilled water used in preparing nutrient solutions for irrigation typically measured 7 to 8 pCi/ml. Such background values were subtracted from all HTO concentration measurements reported in this study. One ml of water plus 14 ml of PCS II was used in liquid scintillation counting

controls, except impinger air sampling controls where a value of water equal to that collected during the sampling period was used (eg 2,500 cc of air collected in 5 mins at 70% RH, 23°C = 21.6 µL of water).

RESULTS AND DISCUSSION

1) Controlled Environment Chamber Performance: Tritium Uptake Kinetics

In early experiments involving tomato plant exposure to tritiated water vapour, steady-state concentrations in the growth chamber were maintained at 5,000 to 7,000 pCi/ml of water whereas in later experiments these concentrations were increased to 10,000 to 15,000 pCi/ml to facilitate tritium-concentration measurement in tomato organs (fruit, foliage) under initial uptake conditions.

Examples of the tritium uptake characteristics of the growth chamber, Conviron Model E15, used during low concentration atmospheric exposures are shown in Appendix 1A and during higher concentration exposures in Appendix 1B. Using a tritium source concentration (wash bottle, Figure 1) of 32 µCi/ml, a delivery flow rate of 100 cc/min and growth chamber exhaust rate of 17 L/min, a steady-state concentration, C_∞ of 6,740 ± 110 pCi/ml ($\bar{x}$ ± 95% confidence interval) was reached, the initial uptake rate constant, k, was 0.138 ± 0.013 hr^{-1} and the initial uptake rate, $C_\infty k$, was 930 pCi/ml/hr At the higher atmospheric tritium exposure concentration (Appendix 1B) the source wash bottle tritium level was 125 µCi/ml, the delivery flow rate 75 cc/min, and the chamber exhaust rate 17 L/min. These conditions resulted in a steady-state concentration of 16,380 ± 550 pCi/ml, an initial uptake rate constant of 0.234 ± 0.024 hr^{-1} and an initial uptake rate of 3,830 pCi/ml/hr based on atmospheric condensate sampling to estimate tritium air concentrations (as in low-level exposure above). Under these conditions the initial uptake rate constant was about double and the initial uptake rate about four times that observed under low-level exposure.

Although air sampling was carried out only by condensate sampling in early experiments direct air sampling using a 2-impinger train was employed in later experiments to estimate tritium atmospheric concentrations, expressed in pCi/ml of water (Appendix IV compares these two methods). This direct air sampling method was developed because of concern for potential lag times in the equilibration of tritium air concentrations with those in atmospheric condensate collected from the chamber refrigeration coils. The chamber tritium uptake concentration curves, measured by these two methods are shown in Appendix 1B. The initial uptake rate constant was 0.318 ± 0.063 hr^{-1}, the initial uptake rate was 4,610 pCi/ml/hr and the steady-state concentration 14,500 ± 980 pCi/ml, as estimated by direct air sampling for 5-minute collection periods. Comparison with the condensate sampling method, showed that the initial rate constants overlapped given the calculated 95% confidence intervals (0.210 to 0.258 versus 0.255 to 0.381), unlike the steady-state

concentrations (13,500 to 15,480 versus 16,030 to 16,930). The mean steady-state concentrations differ by 1880 pCi/ml, that is the condensate estimated air concentrations were 12.9% higher than those obtained from air sampling impingers. More reliable comparisons of the two sampling methods are available in Appendix IV where measurements were replicated (n=5) for each of 6 sampling periods. For a range of air sampling periods of 5 to 30 minutes the tritium concentrations obtained from condensate sampling were 7.9 to 13.3% (mean = 10.3%) higher than those derived from direct air sampling by impinger, (differences significant at 95% level; concentrations by direct air sampling ranged from 18,850 to 19,760 pCi/ml. These differences are consistent with those reported between condensate sampling and collection of water vapour by freeze-trap methods (MacFarlane *et al*, 1979). Throughout these studies tritium chamber atmosphere concentrations have been identified as 'condensate' or 'impinger'.

2) HTO Uptake in Tomato Fruit and Foliage During Atmospheric Exposure to Tritiated Water Vapour

During atmospheric exposure the aerial portions (stem, foliage, fruit, inflorescence) of tomato plants, typically 10 to 14 weeks old, were exposed to tritiated water vapour under two sets of conditions, firstly with the root systems isolated from the atmosphere by root isolation chambers as described in Methods, and secondly without root isolation to determine effects of this variable upon tritium uptake. With root isolation, soil water drainage pumped out of plant pots (Figure 1) was typically at background tritium concentrations ∿7.5 pCi/ml.

The results of exposing tomato plants to tritiated water vapour while the roots were isolated from the atmosphere are shown in Figure 2. The tritium uptake curves for foliage show a very rapid rate of uptake, closely following the rate of the chamber atmosphere, but very unlike the slow rate observed for green fruit. The chamber atmosphere and foliage curves were computer fitted by a non-linear least squares method to the first-order equation $C_t = C_\infty (1-e^{-kt})$ where C_t = tritium concentration in pCi/ml at time t, C_∞ = steady-state tritium concentration in pCi/ml, k = uptake rate constant in hr^{-1} and t = time in hours. This is a first-order kinetic relationship where the uptake rate is proportional to concentration, similar to derivations employed by Kline and Stewart (1969) and Horton et al (1971) which were further developed into mechanistic models by Belot et al (1979) and Garland and Cox (1982). The green fruit uptake data were best fitted by linear regression. The low initial concentrations in green fruit, 20-70 g fresh weight, were in part reflected by the fact that the growth of the fruit was slow (4 to 8 weeks to reach maturity depending on size) which necessitated initiating the uptake experiment with green fruit which had partially developed in clean air. The rapid uptake of tritium in foliage demonstrated the efficient gas exchange properties of foliage compared to the water-conserving functions of fruit.

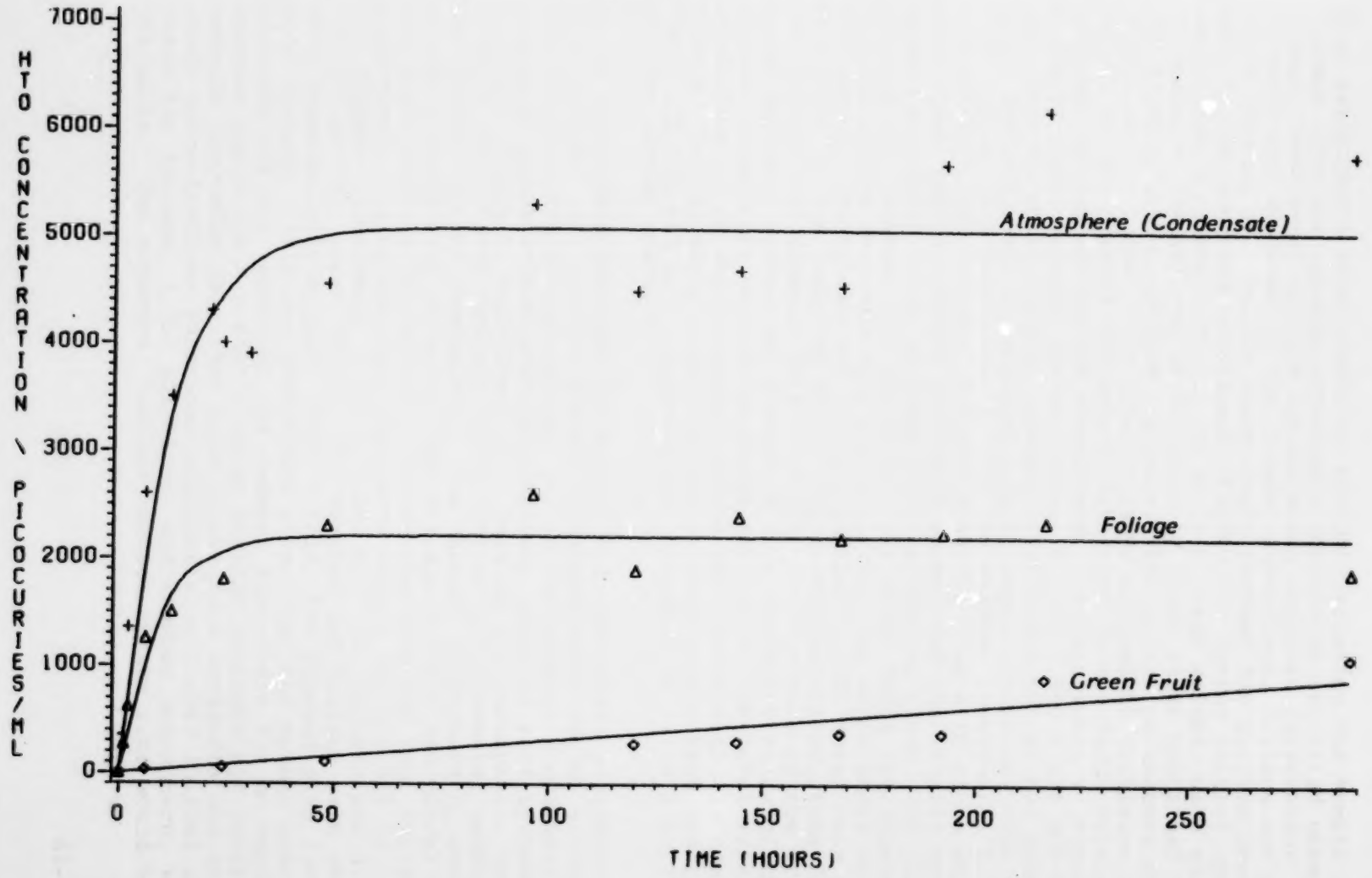


FIGURE 2
HTO UPTAKE IN TOMATO FRUIT AND FOLIAGE
DURING ATMOSPHERIC EXPOSURE TO TRITIATED WATER VAPOUR
WITH ROOTS ISOLATED FROM ATMOSPHERE

159855 RD

When experiments were conducted without isolating the tomato root systems from the chamber atmosphere containing tritiated water vapour, very rapid tritium uptake which followed first-order kinetics was observed in the chamber atmosphere, foliage and green fruit (Figure 3). Unlike green fruit sampled for tritium measurement in the previous experiment (Figure 2), during initial uptake conditions very small fruit, 10 to 15 g fresh weight, were available to minimize the effects of dilution by clean water contained in the fruit prior to tritium exposure. HTO uptake parameters are summarized in Table I for tomato plants exposed to tritiated water vapour in the chamber atmosphere while the root systems were either isolated or exposed to the atmosphere. In experiment 1 (Figure 2) a steady-state tritium concentration, C_∞, of 5,080 ± 430 pCi/ml was reached, while foliage reached 2,210 ± 160 pCi/ml but green fruit did not reach an apparent steady-state condition. The tritium uptake rate constants, k, for the chamber atmosphere, 0.088 ± 0.037 hr^{-1} and foliage, 0.112 ± 0.045 hr^{-1} were in close agreement given the confidence intervals shown, and the initial uptake rates were about twice as great for the chamber alone, 450 pCi/ml /hr, as for foliage, 250 pCi/ml/hr. In green fruit the apparent initial uptake rate was only 2 pCi/ml/hr. The differences between the chamber atmosphere and foliage initial uptake rates were proportional to the observed steady-state concentrations, thus following first-order kinetic relationships. Performance of the tritiated water delivery system and chamber mixing characteristics of the chamber atmosphere were found to be similar to low-level exposure conditions in an empty chamber versus one loaded with six tomato plants. In the empty condition the tritium uptake rate constant was 0.138 ± 0.013 hr^{-1} (Appendix 1A) compared to 0.088 ± 0.037^{-1} for the loaded chamber.

In an experiment designed to measure tritium uptake parameters with exposed root systems during tritiated water vapour exposure, the steady-state concentration, C_∞, in the chamber atmosphere was increased about 2-fold over the previous isolated root system experiment (Table I, experiment 2). Raising the atmospheric concentration facilitated tritium concentration measurements, particularly for initial uptake conditions. The chamber atmosphere C_∞ was 10,180 ± 560 pCi/ml of water, the foliage 5,240 ± 340 pCi/ml and the green fruit 2,520 pCi/ml. Unlike the isolated root system experiment, the uptake rate constants were 4.600 ± 1.919 hr^{-1} for the chamber atmosphere, 0.708 ± 0.288 hr^{-1} for foliage and 0.060 ± 0.037 hr^{-1} for green fruit. The chamber and foliage rate constants were substantially higher, about 6-fold, for exposed than isolated root systems. This was unexpected given that the uptake rate constant for HTO vapour in the chamber atmosphere for an empty chamber was 0.318 ± 0.063^{-1}, using similar impinger air monitoring methods (Appendix 1B). The high uptake rate constants and steady-state concentrations generated substantially higher initial uptake rates for foliage, 3,710 pCi/ml/hr, and chamber atmosphere, 46,830 pCi/ml/hr. The expected foliage initial uptake rate would be

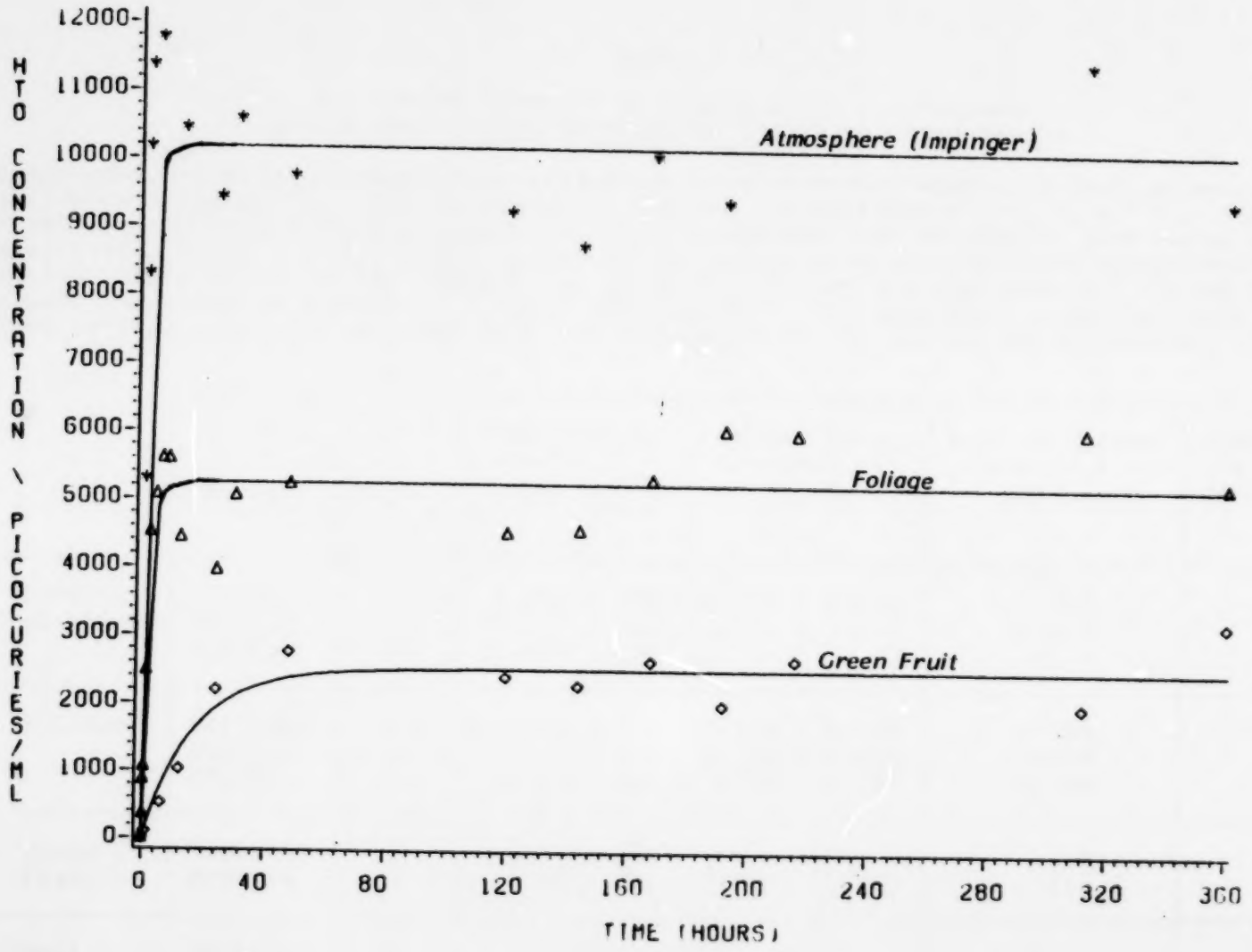


FIGURE 3

HTO UPTAKE IN TOMATO FRUIT AND FOLIAGE DURING **ATMOSPHERIC** EXPOSURE TO TRITIATED WATER VAPOUR WITH ROOTS EXPOSED TO ATMOSPHERE

TABLE I

HTO UPTAKE KINETICS IN TOMATO FRUIT AND FOLIAGE DURING ATMOSPHERIC EXPOSURE TO TRITIATED WATER VAPOUR

Steady-state tritium concentrations and uptake rate constants each expressed as mean value ± 95% confidence interval (σt) were estimated by fitting the equation $C = C_\infty (1-e^{-kt})$. Introduction of HTO into the controlled environment chamber atmosphere was started at time zero, except in experiment 3 where a steady-state concentration (calculated manually) was established before placing tomato plants in the chamber for exposure. Values for experiments 1, 2 and 3 are based on data contained in Figures 2, 3 and 4 respectively. In experiment 1, unlike 2 and 3, the plant root systems were isolated from the atmosphere of the controlled environment chamber.

	Roots	Exp't	Steady-state Conc'n, C_∞ (pCi/ml)	Uptake Rate Constant, k (hr^{-1})	Initial Uptake Rate, $C_\infty k$ (pCi/ml/hr)	Time to Reach 50% Steady-state Conc'n, $t_{1/2}$ (hrs)
Chamber Atmosphere	Isolated	1	5,080 ± 430	0.088 ± 0.037	450	7.9
	Exposed	2	10,180 ± 560	4.600 ± 1.919	46,830	0.2
	Exposed	3	11,980 ± 820	-	-	-
Foliage	Isolated	1	2,210 ± 160	0.112 ± 0.045	250	6.2
	Exposed	2	5,240 ± 340	0.708 ± 0.288	3,710	1.0
	Exposed	3	4,500 ± 340	0.700 ± 0.148	3,150	1.0
Green Fruit	Isolated	1	unattained	-	2	-
	Exposed	2	2,520 ± 300	0.060 ± 0.037	151	11.6
Soil Water	Isolated	1	0	0	0	0

580 pCi/ml/hr (0.112 hr^{-1} x 5,240 pCi/ml) if the same first-order kinetic relationships were to hold for both isolated and exposed root conditions.

A further experiment (Table I, experiment 3) was undertaken to test these observations of substantially increased uptake rate constants and initial uptake rates in exposed versus isolated root systems during atmospheric HTO vapour exposure. Unlike previous Experiments 1 and 2 (Table I) tomato plants, about 12 weeks old, were introduced into a growth chamber already operating at (t=0) steady-state atmospheric tritium concentrations, C_∞ was 11,980 ± 820 pCi/ml (Figure 4). Under these exposure conditions C_∞ for foliage was 4,500 ± 340 pCi/ml and k was 0.700 ± 0.148 resulting in an initial uptake rate, $C_\infty k$, of 3,150 pCi/ml/hr. These values were each similar to those obtained in Experiment 2 (Table I, Figure 3) and suggest that the uptake rate parameters for foliage have not been limited by the introduction of HTO vapour into the chamber at time zero. Yet it is not clear that the difference in rate constants between exposed versus isolated root systems during atmospheric exposure to HTO vapour can be attributed directly to root exposure. In further experiments, involving measurement of both organically-bound and tissue-free water tritium (TFWT) uptake kinetics in tomato (to be described in an upcoming report), TFWT uptake data differ from that presented in Figures 2, 3 and Table I for atmospheric HTO exposure with exposed root systems. With a steady-state concentration value, C_∞, of 4,703 ± 444 pCi/ml and a rate constant, k, of 0.129 ± 0.077 hr^{-1}, the corresponding initial uptake rate was 610 pCi/ml. This k value is interestingly similar to that calculated for isolated root system conditions, k = 0.088 ± 0.037 hr^{-1}. At present it is unclear as to whether the differences between isolated and exposed root systems are attributable to root isolation. It is possible that factors such as leaf age or position and stomatal resistance, which limits gas exchange in foliage, may have limited HTO uptake rates. Other factors which may have affected initial foliar uptake rates include variable greenhouse conditions present during seedling propagation and the duration of plant acclimation prior to HTO exposure. It is recommended that the uptake rate constants and initial uptake rates presented in Table I be taken as a range of values, without necessarily attributing differences to root isolation and exposure.

3) HTO Uptake in Tomato Fruit and Foliage During Irrigation with Tritiated Water

To determine the relative importance of tritium uptake parameters in atmospheric versus soil water tritium exposure six tomato plants, about 12 weeks old, were grown for 17 days during which they were irrigated with nutrient solution containing 20,420 pCi/ml HTO. The growth chamber air change rates were increased from 0.5 to 23 changes/hour to minimize atmospheric tritium concentrations caused by transpiration of tritiated irrigation water. Light intensities, temperature and relative humidity regime, and carbon dioxide levels were maintained as in previous atmospheric tritium exposure experiments (see Methods). At the initiation of the experiment (t = 0) the isolated root systems of each plant were flushed three times

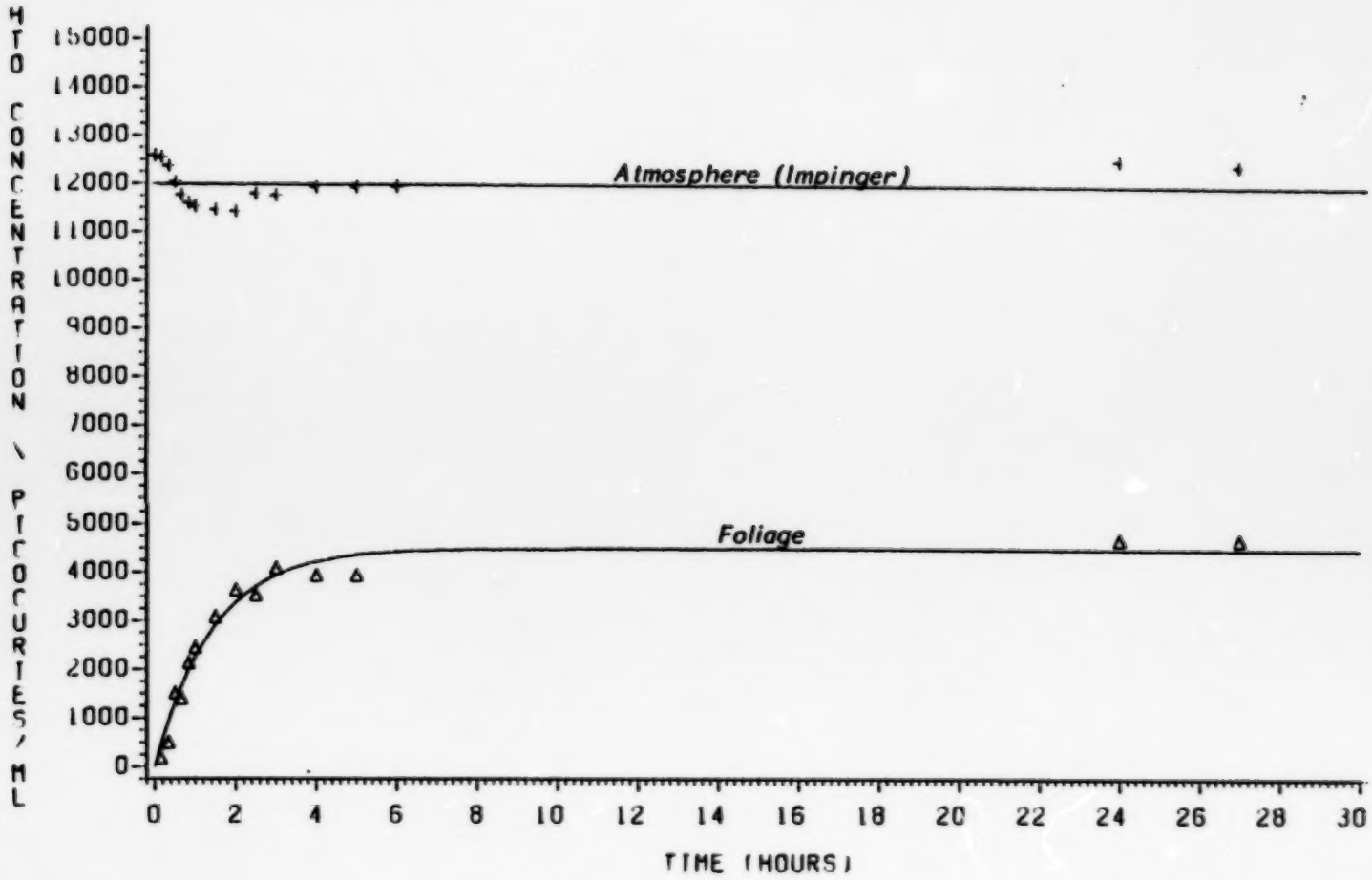


FIGURE 4
TOMATO FOLIAGE HTO UPTAKE DURING EXPOSURE TO A
STEADY-STATE CONCENTRATION OF TRITIATED WATER VAPOUR
WITH ROOTS EXPOSED TO ATMOSPHERE

159857 RD

with 8 L of tritiated nutrient solution to displace the background-tritium soil water. The soil water content was about 200%, (wt water/dry wt soil) equivalent to 4 L per pot.

The HTO uptake rate curves for soil water, atmosphere, foliage and green fruit are shown in Figure 5. The uptake rate constants were similar, 0.023 ± 0.006 to 0.035 ± 0.004, and the initial uptake rates similar for soil water, 450 pCi/ml/hr, green fruit, 345 pCi/ml/hr and foliage, 480 pCi/ml/hr, but lower for chamber atmosphere, 120 pCi/ml/hr, as measured by impinger air sampling (Table II).

The highest steady-state concentration, C_∞, was reached in soil water, 19,430 ± 500 pCi/ml, close to the level supplied in irrigation water, 20,420 pCi/ml. Three plant root systems harvested at the completion (408 hrs) of this experiment were 19,000 ± 1,030 pCi/ml. In decreasing concentration, the C_∞ value for green fruit was 15,000 ± 1,170 pCi/ml for foliage 13,600 ± 440 pCi/ml and for chamber atmosphere 4,440 ± 440 pCi/ml. The atmosphere concentration in this experiment was about 23% of the soil tritium concentration considerably improved over the 62% previously reported using 0.5 air changes/hr (Ontario Hydro Research Division Report 80-52-K).

Comparison of foliage uptake rate constants from tritiated water irrigation (Table I) with those from atmospheric HTO vapour exposure (Table II) during similar root-isolation conditions shows values about four times higher during atmospheric exposure. Green fruit rate constants cannot be compared because under atmospheric exposure the fruit apparently did not reach a steady-state condition. The foliage initial-uptake rate was higher during irrigation water than for the atmospheric exposure experiment with isolated root systems, 480 pCi/ml/hr versus 250 pCi/ml/hr. If tritiated irrigation water rate parameters for isolated roots are compared to the atmospheric exposure rate parameters for exposed roots, then the initial rate constants differ by 20-fold for foliage, 0.035 ± 0.004 hr^{-1} versus 0.700 ± 0.288 hr^{-1} and 3-fold for green fruit, 0.023 ± 0.006 hr^{-1} versus 0.060 ± 0.037 hr^{-1}.

Clearly the above tomato uptake rate constants are specific values for a particular integrated soil-plant system. If clean soil-water had been displaced more rapidly through the use of hydroponic culture techniques, then true rate constants for the plant system alone may have been obtained. They likely would have been substantially larger, even approaching or exceeding those corresponding rate constants obtained under atmospheric exposure conditions. In contrast, under field conditions it is unlikely that natural soils at field water holding capacity would have exceeded the rate constants given here for a common horticultural artificial soil mixture (Redi-Earth). It appears from comparison of the rate constants provided in Table II that the rate of soil water displacement, expressed in terms of the tritium uptake rate constant, would be the rate-limiting step

TABLE II

HTO UPTAKE KINETICS IN TOMATO FRUIT AND FOLIAGE DURING IRRIGATION WITH TRITIATED WATER

Steady-state tritium concentrations and initial uptake rate constants (mean ± 95% confidence interval) were estimated by fitting data to the equation $C = C_\infty (1-e^{-kt})$. Plant root systems were isolated from the controlled environment chamber atmosphere (see Figure 1) and drainage from the isolated roots was used to measure tritium soil water concentrations. Tritium uptake rate curves are shown in Figure 5.

	Steady-state Conc'n, C_∞ (pCi/ml)	Uptake Rate Constant, k (hr^{-1})	Initial Uptake Rate, $C_\infty k$ (pCi/ml/hr)	50% Steady-state Conc'n, $t_{1/2}$ (hrs)
Chamber Atmosphere	4,440 ± 440	0.027 ± 0.009	120	25.7
Foliage	13,600 ± 440	0.035 ± 0.004	480	19.8
Green Fruit	15,000 ± 1,170	0.023 ± 0.006	345	30.1
Soil Water	19,430 ± 500	0.023 ± 0.002	450	30.1

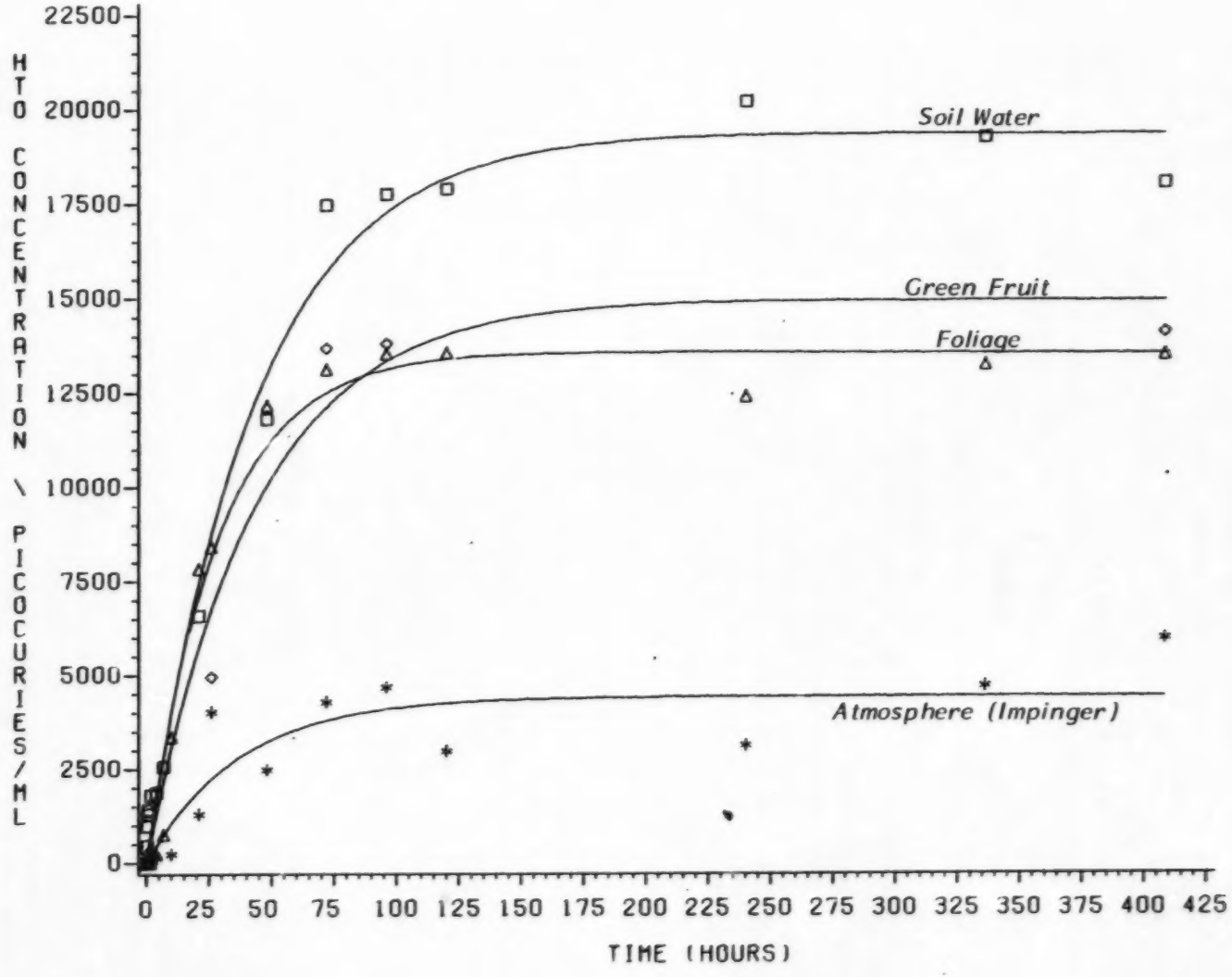


FIGURE 5
HTO UPTAKE IN TOMATO DURING IRRIGATION
WITH TRITITED WATER AND ROOTS ISOLATED FROM ATMOSPHERE

159858 RC

in tritiated water uptake by the various plant organs (foliage, fruit, roots). It should be explained that the soil water displaced in these experiments was primarily that held in the soil pores by capillary forces, and which is normally available to the plant in the range of suction potentials from 10 to 1500 kPa, with 10 kPa being near field capacity, depending on soil type. Other soil water fractions include the free-draining or gravitional water which is very readily displaced if present and hydroscopic water, usually a small fraction, tightly bound to soil particles and unavailable to plants because of suction potentials greater than 1,500 kPa (Brady, 1974).

4) HTO Concentration Ratios of Tomato Fruit and Foliage Relative to Exposure Source

In Table III, comparison is made of the concentration ratios calculated from steady-state concentrations of foliage, green fruit and ripe fruit relative to the HTO exposure levels in the chamber atmosphere and soil water, as measured in pot drainage. With tritiated water vapour exposure in the chamber atmosphere the concentration ratio of foliage to atmosphere, expressed in equivalent concentration units, pCi/ml of water, ranged from 0.38 to 0.51 with a mean value of 0.45 for 4 experiments, 2 with and 2 without root isolation. These values were within the range of ratios reported for other species (Kline & Stewart, 1974; Koranda & Martin, 1973; Belot, 1979; IAEA, 1981). There was no apparent difference between experiments with and without root isolation, suggesting that foliage parameters controlled the attainment of steady-state conditions. Except as noted, variability in these ratios is less than ±15% based on the 95% confidence intervals reported for steady-state concentrations in Tables I and II (this report) and calculated for data given in Table III (Ont Hyd Res Div Report No 80-52-K). In ripe fruit the concentration ratios relative to atmosphere were the same for small (38 - 51 mm) and medium (51 - 57 mm) fruit, 0.18 compared to large (57 - 70 mm) fruit, 0.19. For small green fruit, with exposed root systems, the fruit/atmosphere ratio was 0.25 and for isolated root systems, 0.36 where variability of the latter ratio was about ±30%.

It is expected that the observed concentration ratios for fruit and foliage, 0.18 to 0.51, are substantially less than unity due to the diluting effects of clean soil water moving up in the transpiration stream, the pathway whereby water moves through the vascular system of the roots, stem and foliage before evaporating from the stomata (pores) contained in the foliage. The water supplied by the soil was the major source of water necessary for physiological functions in the plant (Slayter, 1967) and resulted in the net efflux of substantial amounts of water in each tomato plant, about 1 to 1.5 L/day. Clearly the uptake of tritiated water vapour by foliage from the atmosphere must have proceeded by an inward diffusion process, against the large efflux of water vapour from the transpiration stream.

TABLE III

HTO CONCENTRATION RATIOS OF TOMATO FRUIT AND FOLIAGE RELATIVE TO EXPOSURE SOURCE

HTO in tissue-free water was measured in tomato foliage and green and ripe fruit grown in either tritiated atmospheric moisture or irrigation water until steady-state conditions were reached. During atmospheric-HTO exposure soil-water HTO concentration was maintained at background, 7.5 pCi/ml. During HTO-irrigation the atmospheric-HTO concentration was 9,680 ± 2,010 pCi/ml in Experiment 5 and 4,440 ± 440 pCi/ml in Experiment 6. Concentration ratios were calculated from data shown in Table I for Experiments 1, 2 and 3, from Table II for Experiment 6 and from Table III (Ont Hyd Res Div Report No 80-52-K) for Experiments 4 and 5.

Variability in concentration ratios was less than ±15% of values shown, except Exp't 1 where green fruit/atmosphere was about 30%, based on statistical data contained in Tables I and II (this report) and Table III (Report 80-52-K).

HTO Exposure Source	Roots	Exp't	HTO Concentration Ratios				
			Foliage/ Atmosphere	Green Fruit/ Atmosphere	Ripe Fruit/ Atmosphere	Foliage/ Soil Water	Green Fruit/ Soil Water
Atmospheric Fumigation	Isolated	1	0.44	-	-		
	Isolated	4	0.45	0.36	0.19[1]		
	Exposed	2	0.51	0.25	0.18[2]		
	Exposed	3	0.38	-	-		
Irrigation Water	Isolated	5	1.50	1.48		0.83	0.82
	Isolated	6	3.06	3.38		0.70	0.77

[1] Small and medium sized fruit values similar
[2] Large fruit

Isotopic discrimination, favouring uptake of HOH over HTO, as demonstrated by McFarlane (1976) and Garland & Ameen (1978) in lettuce, barley and maize leaves also may have contributed to the low foliar/atmosphere concentration ratios for tomato. In the type of experimental procedure followed for tomato, it was not possible to separate the relative contribution of isotopic discrimination and dilution by soil water to the lowering of foliar and fruit steady-state concentrations below atmospheric levels.

With tritiated water irrigation the concentration ratio of foliage to soil water was 0.70 during which the foliage/atmosphere ratio was 3.06 (Table III, Exp't 5). In a previous experiment (Exp't 6) the foliage/soil water ratio was 0.83 and the foliage/atmosphere ratio 1.50. The latter experiment had been carried out with the growth chamber operating at 23 air changes/hr, the maximum rate attainable, while the earlier experiment (Exp't 5) involved 0.5 air changes/hr, similar to atmospheric exposure conditions. The purpose of operating the chamber at maximal air changes was an attempt to establish background-tritium concentration in the atmosphere. The differences in the concentration ratios given for the two tritiated irrigation water experiments suggest that the effects of reduced tritium atmospheric levels was to reduce the contribution of tritiated irrigation water towards a foliage/soil water ratio approaching unity. The values given for green fruit/soil water ratios suggest the same pattern, although the variability (±15%) in each case precludes attaching any statistical significance to the differences in foliage/soil water and fruit/soil water ratios.

In Table III atmospheric HTO exposure values were not given for the foliage and green fruit/soil water ratios because the soil water concentration was zero once the background of 7.5 pCi/ml was subtracted.

5) HTO Desorption from Harvested Ripe Tomato Fruit

This experiment was designed to test whether or not after harvest ripe fruit preferentially lost tritiated water by desorption in a clean atmosphere. Such information would be useful in predicting the tritium desorption rates and radiation dose in ripe fruit grown in proposed waste-heat utilization greenhouses at nuclear generating facilities. In Figure 6, fruit HTO concentrations are shown for periods up to 364 hours following harvest for three commercial fruit size classes grown from early fruit-set to maturity in tritiated water vapour. The size classes were based on the Ontario Ministry of Food and Agriculture Marketing grades where small fruit were 38 to 51 mm diameter, medium 51 - 57 mm, large 57 - 70 mm and extra large greater than 70 mm. Fruit was harvested on four separate occasions (1 harvest of large fruit, 3 harvests of small and medium fruit) and placed in a laboratory at temperatures of 24 ± 1°C with relative humidities ranging from 36 to 56%. It should be noted that large fruit had been grown at high atmospheric tritium concentrations (~10,000 pCi/ml) and small and medium fruit at low

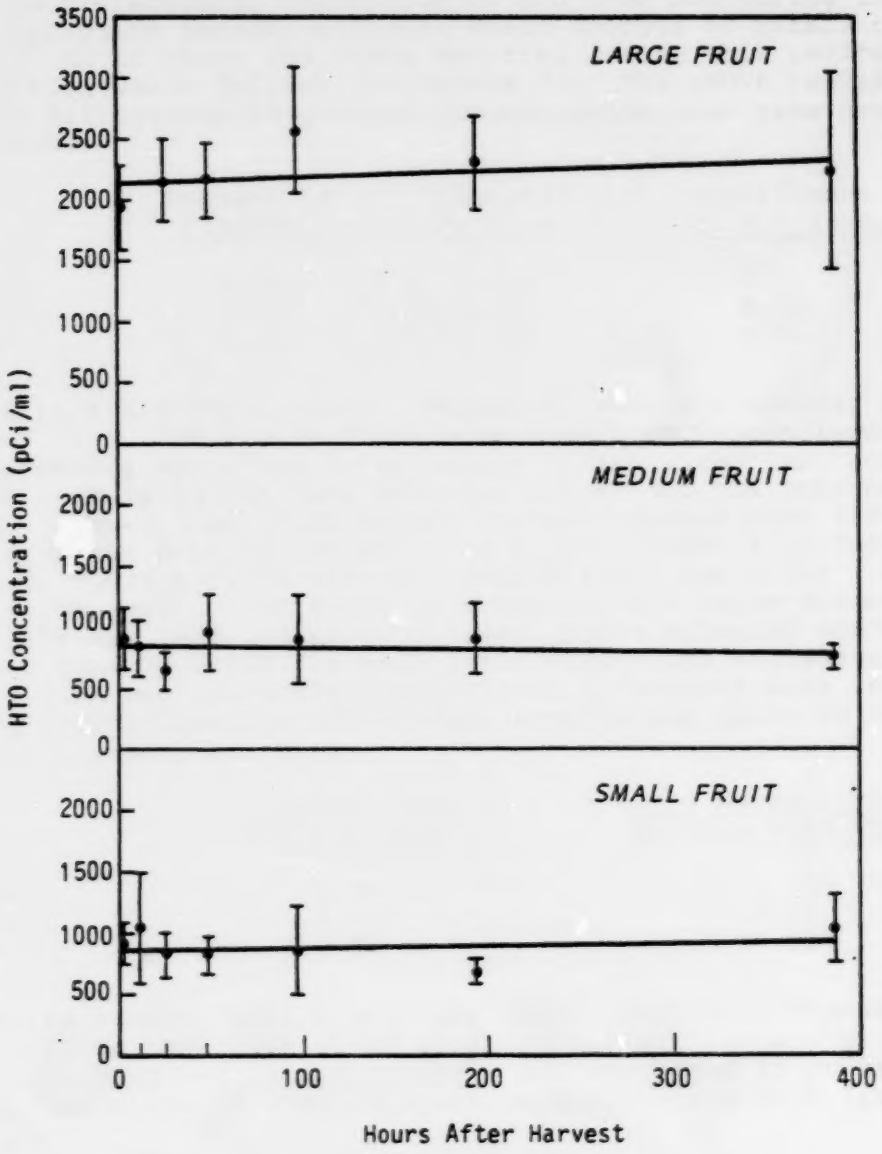


FIGURE 6

HTO DESORPTION IN HARVESTED TOMATO FRUIT

Mean concentrations ±1 Standard Deviation are shown for n = 4 to 6 Replicates, except t = 0, where n = 8

159177 RD

concentrations (∿5,000 pCi/ml). Statistical analysis (ANOVA) of the fruit HTO concentration data contained in Figure 6 demonstrated that at the 95% probability level the HTO concentrations did not differ with time for each of the three fruit size classes. Clearly there was no preferential desorption of HTO over HOH during the 364-hour desorption period, although small amounts of water, both HOH and HTO up to about 10%, were desorbed during this period (see discussion which follows for Figure 7). The ANOVA results for testing differences in tritium concentration over time are outlined below:

	Degrees of Freedom	Computed F-Value	Significant at 95% level
Large fruit	1,36	0.664	N.S.
Medium fruit	1,34	0.231	N.S.
Small fruit	1,40	0.246	N.S.

The losses in fruit fresh weight (Figure 7) were due chiefly to the loss of water. For each fruit size class, ANOVA confirmed that there were significant fresh weight losses with time, and subsequent regression analyses provided values for the intercept and for the slope. The fresh weight losses together with 95% confidence intervals were estimated to be 0.016 ± 0.004 %/hr for large fruit, 0.025 ± 0.004 %/hr for medium fruit and 0.033 ± 0.008 %/hr for small fruit. The trend here was for the large diameter fruit to lose the least water over time. Calculation of mean volumes and surface areas for each fruit size class suggested that water loss was inversely proportional to surface area rather than being proportional to the surface area/volume ratio as expected from physical principles.

	% Water loss / Surface area	% Water loss / Surface area/volume
Large fruit	2.46	0.186
Medium fruit	2.62	0.240
Small fruit	2.82	0.287

ANOVA results showing that there were significant differences at the 95% level in the fresh weights of fruit with time are given below. Subsequent regression analysis was performed to provide values for the slope (% loss in fresh weight/hr) for each fruit size class.

	Degrees of Freedom	Computed F-value	Significant at 95% level	Slope (% loss/hr)
Large fruit	1,36	74.320	Yes	0.016
Medium fruit	1,34	251.362	Yes	0.024
Small fruit	1,40	81.040	Yes	0.033

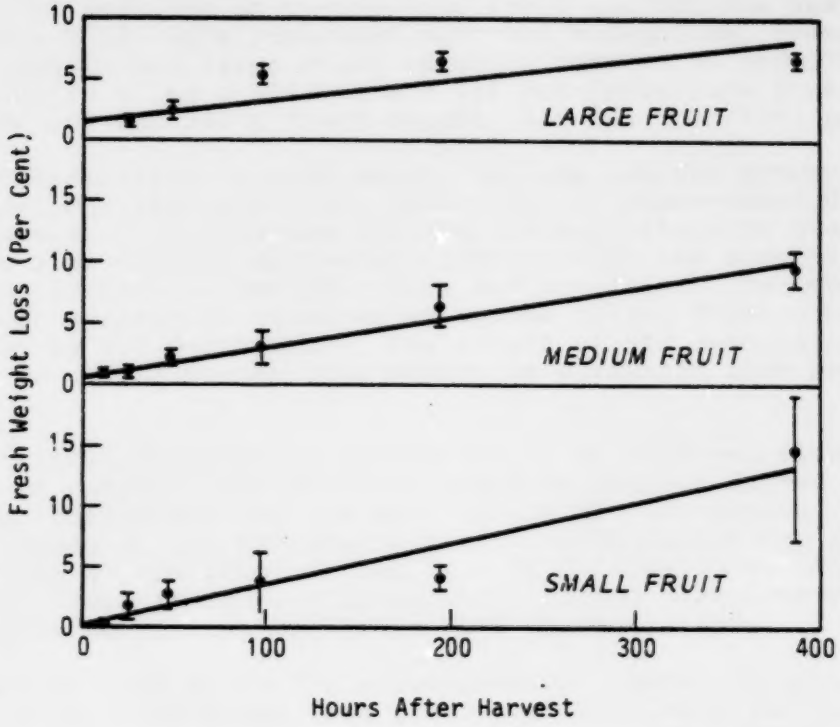


FIGURE 7

FRESH WEIGHT LOSS
AFTER TOMATO FRUIT HARVEST

Mean fresh weight losses ±1 Standard Deviation are shown for n = 4 to 6 Replicates, except for t = 0, where n = 8

6) On-The-Vine HTO Concentration Changes in Tomato Fruit and Foliage

Fifteen-week old plants bearing green fruit of various sizes were placed in the growth chamber, the roots isolated, then irrigated with 18,340 ± 2,660 pCi/ml tritiated water containing nutrients. After 20 days the tritiated water irrigation was terminated and the pots flushed with 8L of tap water per pot, on 3 successive occasions, to initiate an on-the-vine fruit and foliage desorption experiment. Fruit were separated into two categories, green fruit of small, medium and large sizes weighing between 40 and 370 g fresh weight ($\bar{x}$ = 164 ± 98(SD), n = 21) and large ripe fruit weighing between 100 and 340 g fresh weight ($\bar{x}$ = 225 ± 75(SD), n = 16).

The HTO concentration in soil water, foliage and the atmosphere declined rapidly indicating HTO desorption by clean-water displacement (Figure 8). In contrast the HTO concentrations in green fruit of various size classes decreased slowly during the same 696-hour clean-water irrigation period. This was the likely consequence of the diluting effects of clean-water uptake during fruit growth, rather than by HTO desorption. The extent of HTO desorption, if any, in green fruit on-the-vine cannot be estimated from present data.

The normal fruit development period was 30 to 45 days, thus at 240 hours, about one-half of the fruit fresh weight was formed during clean-water irrigation and one-half during HTO irrigation. As shown in Figure 8, the HTO concentration at 240 hours was about one-half that at the initial time (t = 0). Likewise at 696 hours most of the green fruit were formed only during clean-water irrigation resulting in near background HTO concentrations.

The pattern of ripe fruit HTO concentration change, if any, was unclear. Ripe fruit (large size class) were available for harvest only at initial time and on three sampling periods near the end of the experiment, 408, 576 and 697 hours (Figure 8). The low initial HTO concentration of ripe fruit $\bar{x}$ = 4,980 ± 760 pCi/ml compared to green fruit 17,350 ± 310 pCi/ml was attributed to partial development of ripe fruit prior to the 20-day HTO irrigation period. If a similar developmental regime were followed by ripe fruit sampled at 408, 576 and 696 hours, or if little growth had occurred during the clean-water irrigation period then the data would suggest no substantial HTO desorption. Data were not available on the amount of ripe fruit growth. An alternative explanation is that the HTO concentrations in ripe fruit sampled at 408, 576 and 696 hours resulted from the dilution of initially high HTO levels by clean water during growth, as suggested for green fruit. Excepting ripe fruit, HTO desorption followed first-order kinetics, thus the data was fitted by non-linear least squares to the equation $C_t = C_i\ (e^{-kt})$ where C_i is the initial tritium concentration in pCi/ml (at t=o), k is the desorption rate constant and the other parameters are as defined earlier for uptake.

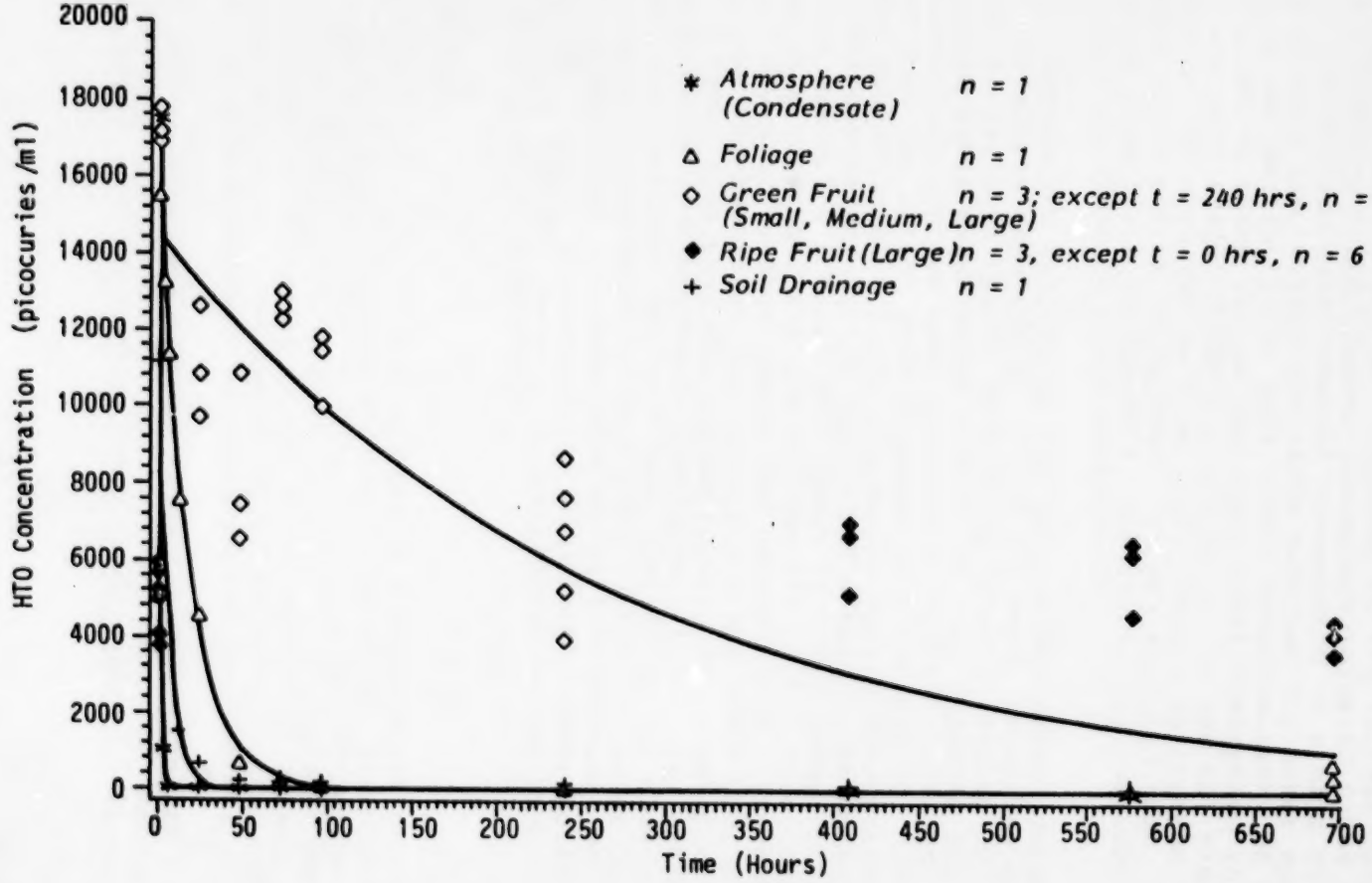


FIGURE 8

ON -THE-VINE CHANGES IN HTO CONCENTRATIONS
OF FRUIT AND FOLIAGE WHEN HTO IRRIGATION
WAS FOLLOWED BY CLEAN WATER

The initial tritium concentration, C_i, was highest in soil water 17,500 ± 30 pCi/ml, followed by foliage at 15,600 ± 440 pCi/ml green fruit at 14,470 ± 1,960 pCi/ml, the chamber atmosphere at 11,190 ± 430 pCi/ml (Table IV). The soil water desorption rate was large, 0.950 ± 0.011 hr^{-1} compared to the foliage value of 0.056 ± 0.004 hr^{-1} and chamber atmosphere value of 0.149 ± 0.021 hr^{-1}. The foliage rate constant corresponds to a half-time, $t_{1/2}$, value of 12.4 hrs, within the range reported for other species (Koranda and Martin, 1973; IAEA, 1981; Krishnamoorthy et al, 1979). In contrast to these values the green fruit desorption rate constant was very small, 0.004 ± 0.001 hr^{-1}. It was not clear if any desorption had occurred in ripe fruit, thus a desorption rate constant could not be calculated. It was expected that, like harvested ripe fruit in Figure 6, little, if any, desorption would take place in ripening fruit on-the-vine.

Kinetic parameters calculated for tritium desorption during clean-water irrigation experiments (Table IV) were, excepting green fruit values, larger than uptake parameters obtained during tritiated irrigation water experiments (Table II). As expected the chamber atmospheric desorption rate constant of 0.159 ± 0.021 hr^{-1} was larger than the uptake rate constant of 0.027 ± 0.009 hr^{-1} due to chamber operation at 0.5 air changes/hr during desorption compared to 23 air changes/hr during uptake. The soil water desorption rate constant was 0.950 ± 0.011 hr^{-1} about 50 times larger than the uptake rate constant, and the foliage desorption rate constant 0.056 ± 0.004 hr^{-1}, about twice the uptake value. In contrast the desorption rate constant for green fruit was 0.004 ± 0.001 hr^{-1}, about 5 times less than the uptake value. It is not clear why the desorption rate constants were generally higher, although the uptake rate constants may have been limited by the slow rate of clean water displacement by tritiated irrigation water during the uptake experiments. Factors such as soil compaction, soil water content at time of initiating the experiment, water holding capacity and factors controlling transpiration may have limited the rate of soil water displacement.

CONCLUSIONS

Tritiated water (HTO) uptake and desorption kinetics have been measured in the tissue-free water tritium (TFWT) fractions of tomato fruit and foliage exposed to HTO, to gain a better understanding of tritium behaviour in vegetation surrounding nuclear facilities. This information is useful in the construction of dynamic predictive models describing the environmental behaviour of tritium and in more definitive assessment of radiological dose to man.

Tomato plants have been exposed to HTO under controlled conditions of temperature, light intensity, relative humidity, carbon dioxide concentration, and irrigation. Two basic experimental regimes

TABLE IV

HTO DESORPTION KINETICS IN TOMATO FRUIT AND FOLIAGE WHEN HTO IRRIGATION WAS FOLLOWED BY CLEAN WATER

Tomato plants bearing fruit (105 days old) and with roots isolated were irrigated with 18,340 ± 2,660 pCi/ml tritiated water for 20 days followed by clean-water irrigation for 29 days (Fig 8). The tritium desorption rate constants, k, and initial concentrations, C_i, at t = 0, mean ± 95% confidence interval, were estimated by fitting data to the equation $C_t = C_i\ (e^{-kt})$ using a non-linear least squares method.

	Initial Conc'n, C_i (pCi/ml)	Desorption Rate Constant, k (hr^{-1})	Initial Desorption Rate, $C_i k$ (pCi/ml/hr)	Time to Reach 50% of Initial Conc'n, $t_{1/2}$ (hrs)
Chamber Atmosphere	11,190 ± 430	0.159 ± 0.021	1,780	4.4
Foliage	15,600 ± 440	0.056 ± 0.004	870	12.4
Green Fruit	14,470 ± 1,960	0.004 ± 0.001[1]	54	173.0
Soil Water	17,500 ± 30	0.950 ± 0.011	16,610	0.7

[1] The rate constant for green fruit likely reflects the dilution of initially high HTO concentrations by clean water uptake during growth, rather than HTO loss.

have been followed in these studies: 1) plants with isolated and exposed root systems were fumigated with atmospheric tritiated water vapour during clean-water irrigation and 2) plants having isolated root systems were irrigated with tritiated water.

The conclusions drawn from these experiments were as follows:

1) HTO uptake and desorption into tissue-free water fractions of tomato foliage and most fruit followed first-order kinetics, ie uptake or desorption rates were proportional to exposure concentrations. For uptake, data were fitted by a non-linear least squares procedure to the equation $C_t = C_\infty (1-e^{-kt})$, where C_t = HTO concentration at time t in pCi/ml of water, C_∞ = steady-state concentration in pCi/ml and k = uptake rate constant in hr^{-1}. Likewise, most desorption data fitted the equation $C_t = C_i (e^{-kt})$, where C_i = the initial HTO concentration in pCi/ml of water and k = desorption rate constant. Initial uptake rates, $C_\infty k$, and the time to reach 50% of the steady-state concentration, $t_{1/2}$ = 0.693/k, are provided in Tables I, II, III and IV. The tomato fruit and foliage tissue-free water tritium contents were 91.5 ± 2.1% (n=13) and 94.0 ± 3.7% (n=16) of their respective weights.

2) With both atmospheric HTO and root HTO exposure by irrigation, tritium uptake was more rapid in foliage than green fruit. The foliage uptake rate constants for atmospheric HTO exposure ranged from 0.112 ± 0.045 hr^{-1} (roots isolated; ± 95% confidence interval) to 0.708 ± 0.288 hr^{-1} (roots exposed) and the green fruit uptake rate constant 0.060 ± 0.037 hr^{-1}, (roots exposed). With tritiated irrigation, the foliage uptake rate constant was 0.035 ± 0.004 hr^{-1} and green fruit 0.023 ± 0.006 hr^{-1} (Tables I & II). It is important to note that under field or greenhouse conditions the rate constants obtained during tritiated water irrigation could increase or decrease substantially depending on prior tritium concentrations in the soil and soil water turnover rates.

3) The average steady-state HTO concentration ratio for foliage/atmosphere was 0.45 with values ranging from 0.38 to 0.51 in 4 experiments. There was no evidence that root isolation affected these values. The average value of 0.45 was in very close agreement with the ratio of 0.5 used by the US Nuclear Regulatory Commission in their specific activity model. In two of the four experiments where green fruit HTO concentrations were measured, the green fruit to atmosphere steady-state concentration ratio was 0.19 (range 0.18 to 0.19) and the ripe fruit/atmosphere ratio was 0.31 (range 0.25 to 0.36). The green fruit/atmosphere and ripe fruit/atmosphere ratios did not differ given the variability in the data (Table III).

4) In two tritiated water irrigation experiments, the HTO concentration ratio at steady-state for foliage/soil water was 0.70 to 0.83 and the green fruit/soil water ratio was 0.77 to 0.82. During these measurements the corresponding ratio of foliage/atmosphere was 1.50 to 3.06 and green fruit/atmosphere a similar 1.48 to 3.38. These data suggests that a lower atmospheric HTO concentration acted to reduce the foliage and fruit concentrations during tritiated water irrigation.

5) No significant (95% probability level) decrease in HTO concentration was observed over 364 hours in ripe fruit of three commercial size classes: small, medium and large (Figure 6). Over the same period there was significant water loss from each fruit size class indicating proportional loss of HTO and HOH. The rates of loss were 0.016%/hr for large fruit, 0.024%/hr for medium fruit and 0.33%/hr for small fruit (Figure 7). The desorption and uptake data have important implications for tomatoes grown in nuclear waste-heat greenhouses or in field produce grown in the environs of nuclear facilities. The present data suggests that during growth produce would reach a maximum HTO concentration of about 45% of the atmosphere or about 85% of the soil water whichever is higher. If produce has water-conserving properties similar to tomato fruit (eg apples, pears) then preferential HTO desorption would not be expected following harvest. Rapid equilibration, $t_{1/2}$ = 1 to 6 hours, with the atmosphere would be expected for foliar produce unlike fruit.

6) The rate constant for on-the-vine green fruit tritium desorption during clean water irrigation was small, 0.004 ± 0.001 hr^{-1}, and for ripe fruit a rate constant could not be estimated. In contrast, the foliage desorption rate constant was 0.056 ± 0.004 hr^{-1} and 0.950 ± 0.011 for the displacement of tritiated soil water by clean water (Table IV).

Approved:

D. Harrison
Manager
Chemical Research Dept

Submitted:

F.S. Spencer
Biologist
Biological Research Section

FSS:aba:rdm

REFERENCES

Belot, Y., D. Gauthier, H. Camus and C. Caput. (1979) Prediction of the Flux of Tritiated Water from Air to Plant Leaves. Health Physics 37, 575-583.

Brady, N.C. (1974) The Nature and Properties of Soils. MacMillan Publishing Co., N.Y. p 192.

Eisenbud, M. et al (1979) Tritium in the Environment - NCRP Report #62 In: Behaviour of Tritium in the Environment. IAEA, Vienna.

Garland, J.A. and M. Ameen (1979) Incorporation of Tritium in Grain Plants. Health Physics 36, 35-38.

Garland, J.A. and L.C. Cox (1982) Uptake of Tritiated Water Vapour by Bean Leaves. Water, Air and Soil Pollut. 17 207-212.

Gorman, D.J. (1979) Interim Report, Pickering Greenhouse Project, September 5, Ontario Hydro CSS.

Horton, J.H., J.C. Corey and R.M. Wallace (1971) Tritium Loss from Water Exposed to the Atmosphere. Env. Sci. Technol. 5, 338-343.

IAEA (1981) Tritium in Some Typical Ecosystems. Technical Reports Series No 207. IAEA, Vienna.

Kline, J.R. and M.L. Stewart (1974) Tritium Uptake and Loss in Grass Vegetation which has been Exposed to an Atmospheric Source of Tritiated Water. Health Phys 26, 567-573.

Koranda, J.J. and J.R. Martin (1973) The Movement of Tritium in Ecological Systems. In: A.A. Moghissi and M.W. Carter, eds. Tritium Conf 710809. p 430-455. Messenger Graphics, Las Vegas, Nev.

Krishnamoorthy, T.M. et al (1979) Behaviour of Tritium in the Plant Domain. In: Behaviour of Tritium in the Environment. p 386-403. IAEA, Vienna. STI/PUB/498.

Langhans, R.W., ed (1978) A Growth Chamber Manual. Cornell University Press, Ithaca.

McFarlane, J.C. (1976) Tritium Fractionation in Plants. Environ Expt Bot 16, 9-14.

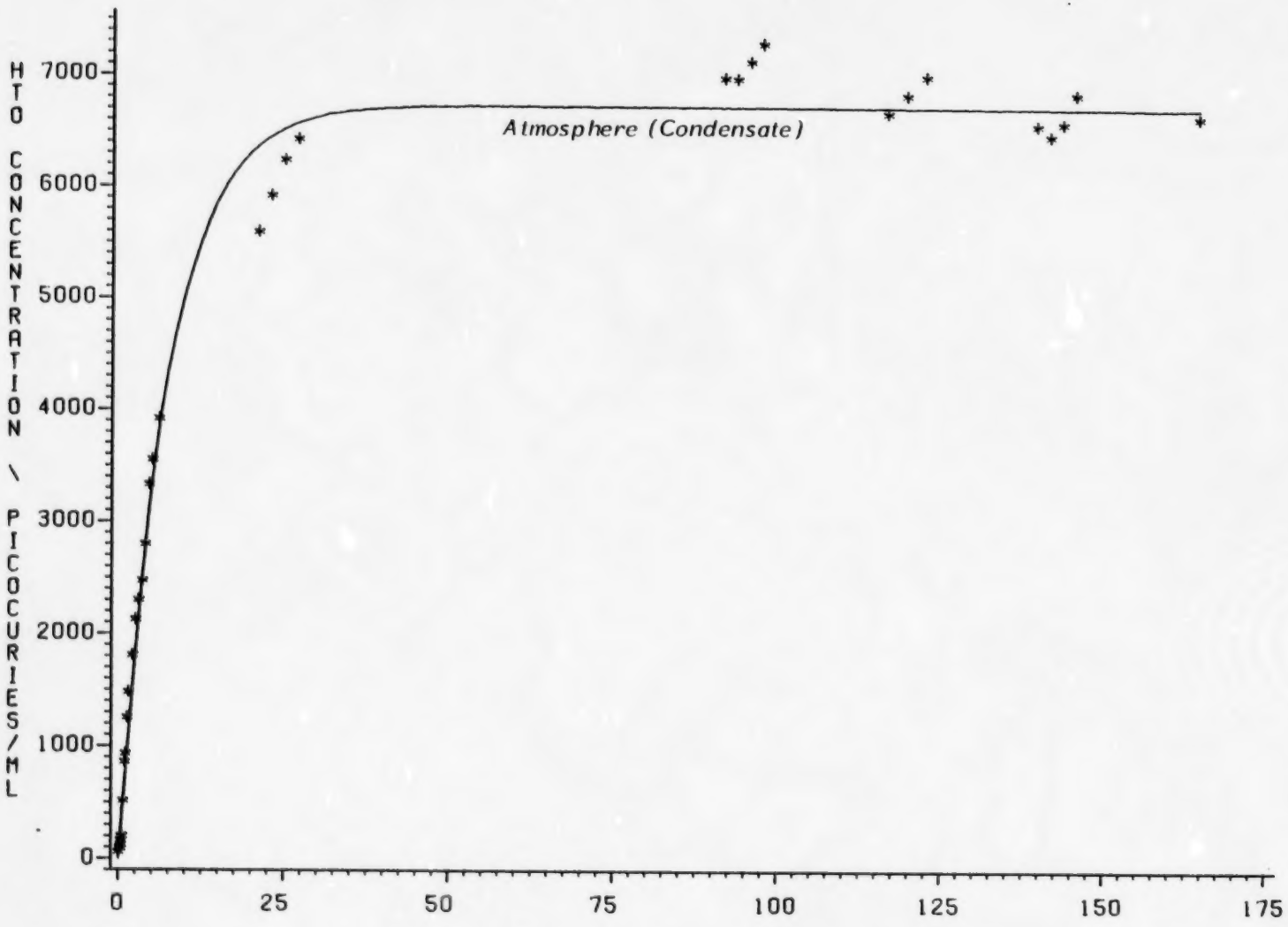
McFarlane, J.C., W.F. Beckert and K.W. Brown (1979) Tritium in Plants. J. Environ Qual 8, 269-276.

Moghissi, A., E.W. Bretthauer and E.H. Compton. Separation of Water from Biological and Environmental Samples for Tritium Analysis. Analytic Chemistry 45, 1565.

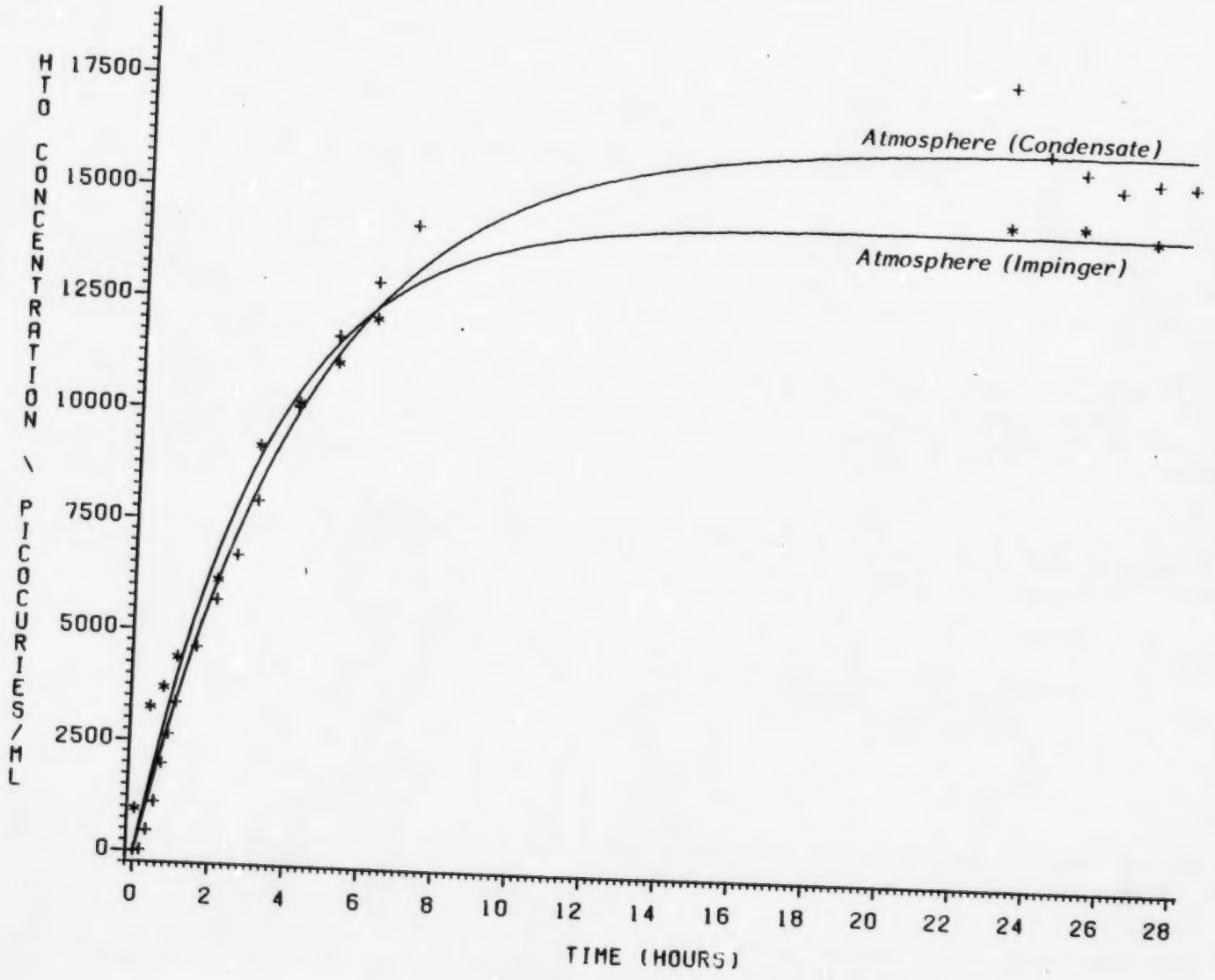
Ontario Hydro Research Division Report No 80-52-K. (1980) Tritium Uptake and Desorption Kinetics in Horticultural Species. Interim Report. F.S. Spencer.

Quisenberry, D.R. (1979). Environemntal Aspects of Tritium. Environ Pollut. 20, 33-43.

Slayter, R.O. (1967). Plant Water Relationships. Academic Press, N.Y.



APPENDIX 1A
CONTROLLED ENVIRONMENT CHAMBER HTO UPTAKE
CHARACTERISTIC FOR LOW-LEVEL EXPOSURE



APPENDIX 1B
CONTROLLED ENVIRONMENT CHAMBER HTO UPTAKE
CHARACTERISTICS FOR HIGH-LEVEL EXPOSURES

APPENDIX II

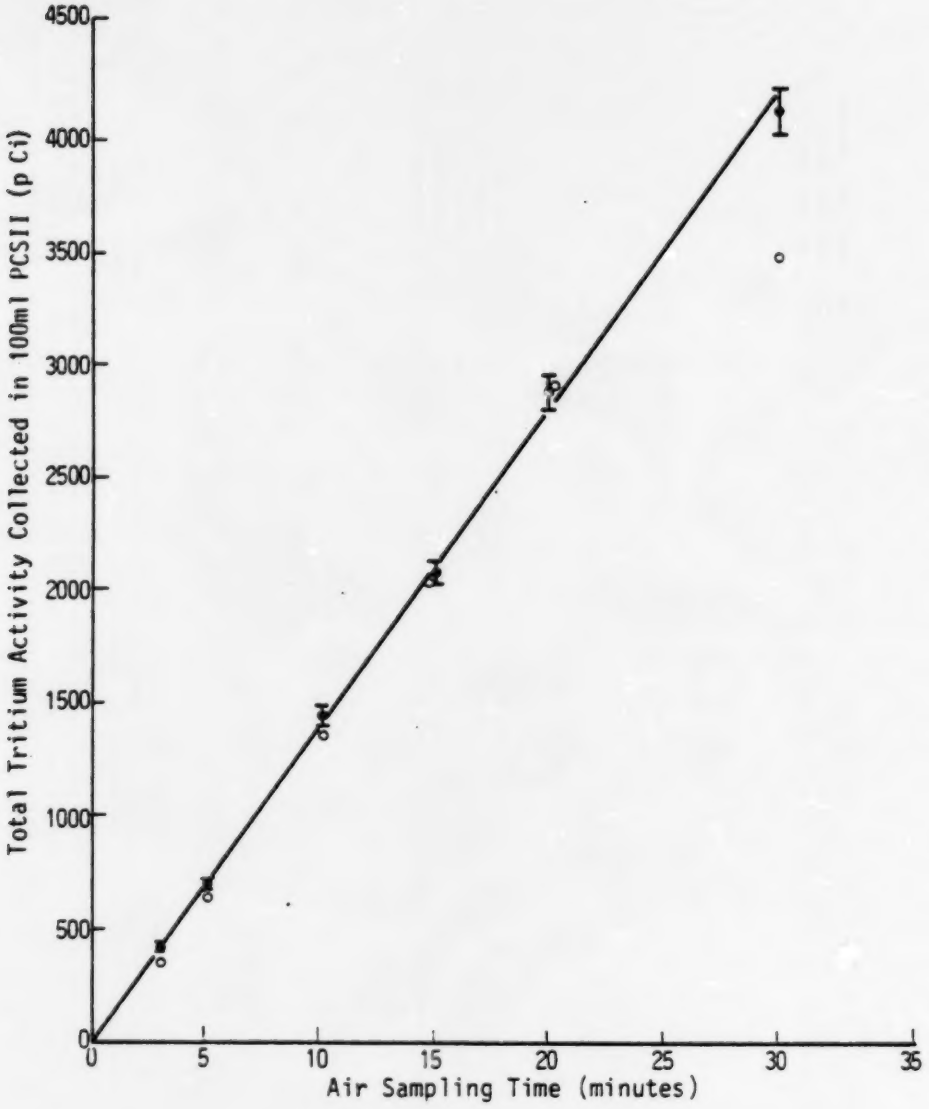
HTO COLLECTION EFFICIENCY WITH A TWO-IMPINGER TRAIN

An air sampling train consisting of two 125 ml glass impingers was used for collecting tritiated water vapour maintained at a steady-state concentration in a CONVIRON E 15 controlled environment chamber. Air was drawn through the sampling train by a downstream Metal Bellows Pump (MB-21). The flow rate of 500 cc/min was controlled by a calibrated Matheson 602 rotameter.

For each measurement 50 ml of PCS II liquid scintillation cocktail (Amersham Corp) was placed in each impinger to sample the tritiated water vapour in the chamber atmosphere. Values shown are means, n = 5, ± standard deviation. Blanks constituted an equivalent amount of distilled water for each collection period (eg for 3 mins, 21.6 µl water collected at 70% RH, 23°C).

Collection Period (mins)	Tritium Activity in 50 ml PCS II		Impinger No 1 Collection Efficiency (% of Total Activity Collected)
	Impinger No 1 (pCi)	Impinger No 2 (pCi)	
3	426 ± 17	3 ± 3	99.3
5	692 ± 22	4 ± 4	99.4
10	1427 ± 30	26 ± 10	98.2
15	2026 ± 56	58 ± 7	97.2
20	2790 ± 84	110 ± 6	96.2
30	3862 ± 83	281 ± 18	93.2

Notes:
1. An alternative collection method (Appendix IV) provided tritium specific activity values in atmospheric condensate about 10% higher than those shown above on a per ml basis.
2. A third impinger, placed in series with the above, collected less than 0.5% of total tritium activity at 15 and 30 minute periods.



APPENDIX III

EFFECT OF SAMPLING TIME ON IMPINGER PERFORMANCE

Two impingers, each containing 50ml PCSII, were used. Total tritium activity values are shown as mean ±1 standard deviation for 5 replicates (●); a glass wool filter was placed in the sampling line for one additional measurement (○).

APPENDIX IV

COMPARISONS OF ATMOSPHERIC CONDENSATE AND IMPINGER AIR SAMPLING FOR MEASUREMENT OF CHAMBER HTO CONCENTRATIONS

Two-way ANOVA was carried out to test whether the two methods for estimating chamber atmospheric tritium concentrations were different at the 95% level. These methods were a two-impinger sampling train described in Appendix II and collection of chamber atmospheric condensate from refrigeration coils which were followed by liquid scintillation counting (See "Methods"). Chamber air was sampled for various time periods (3 to 30 mins) from a chamber operated under steady-state atmospheric tritium concentrations (cf Appendix 1B). Temperature, relative humidity and volume of air sampled were measured. Condensate was sampled in the final 0.5 minute of each air sampling period for comparison.

The two-way ANOVA table below demonstrated that there was significant interaction between the duration of sampling and the method of sampling ($F(1,5)_{\alpha=0.05} = 6.61$). This implied that the differences between the methods varied with the duration of sampling.

	DF	Sum of Squares	Actual F-Value
Method of Sampling	1	47,337,507	236.73
Duration of Sampling	5	14,241,843	14.24
Interaction (Method x Duration)	5	5,290,687	5.29

Because the differences between methods varied with collection period, differences were examined on an individual collection period basis. Estimates of the differences were made by a regression model and those differences found to be significant at the 95% level are shown. Tritium air concentrations in pCi/ml of water are shown as mean values ± 95% confidence intervals for 5 replicates at each collection period. Method difference estimates also include 95% confidence intervals. NS = not significant at 95% level.

Collection Period (mins)	Air Sampling Method: 2-Impingers (pCi/ml)	Air Sampling Method: Condensate (pCi/ml)	Estimate of Difference pCi/ml
3	19,850 ± 1,980	20,530 ± 1,270	NS
5	18,850 ± 1,480	21,370 ± 440	2,520 ± 640
10	19,800 ± 1,260	21,900 ± 370	2,100 ± 540
15	18,940 ± 1,230	20,630 ± 880	1,690 ± 630
20	19,760 ± 1,450	21,950 ± 480	2,190 ± 630
30	18,820 ± 1,050	20,300 ± 330	1,480 ± 460

The condensate sampling method provided values about 10.3% higher than the corresponding air sampling values obtained for 5 to 30 minute collection periods. Normally 5-minute collection periods were used throughout this report where chamber air concentration was sampled by the direct air sampling method.

POOR

QUALITY

ORIGINAL